

### Remarks

In the Office Action mailed September 13, 2004, claims 1-20 were finally rejected under 35 U.S.C. §103(a) as being unpatentable over Hartley et al., U.S. Patent No. 4,853,737 ("Hartley") and, incorporated by reference, Lentz, U.S. Patent No. 4,257,699 ("Lentz") in view of Schlueter, Jr. et al., U.S. Patent No. 5,995,796 ("Schlueter"), Kirk-Othmer, "Elastomers, Polyisoprene to Expert Systems," *Encyclopedia of Chemical Technology*, pp. 16-20, 22-25 (4<sup>th</sup> Ed., Vol. 9, John Wiley & Sons, 1994) ("Kirk"), and Lewis, *Hawley's Condensed Chemical Dictionary*, pp. 437, 1097 (13<sup>th</sup> Ed., Van Nostrand Reinhold Publ., 1997) ("Lewis"). Also, claims 1-22 were finally rejected under 35 U.S.C. §103(a) as being unpatentable over Hartley in view of Schlueter, Blong et al., U.S. Patent No. 5,527,858. ("Blong"), Kirk, and Lewis. These rejections were maintained in the subsequent Advisory Actions.

Hartley, the primary reference cited in the rejection of the claims of the instant application, discloses a fuser roll having an outer layer that comprises cured fluorooelastomer having pendant polydiorganosiloxane segments that are covalently bonded to the fluorooelastomer backbone. DuPont Viton A and Viton B are cited as suitable fluoroelastomer base polymers. Lentz and Schlueter, like Hartley, disclose compositions containing cured fluorooelastomers such as Viton A and Viton B.

Eddy et al., U.S. Patent No. 5,017,432 ("Eddy"), which was cited by the Examiner in the March 5, 2002 Office Action and briefly discussed in the December 16, 2004 Supplemental Advisory Action, discloses a fuser member whose fluoroelastomer fusing surface comprises a VF-HFP-TFE terpolymer containing less than 40 mole% VF, for example fluorooelastomer Viton GF, available from DuPont (column 9, lines 15-21). Elsewhere in the disclosure (column 3, lines 20-58; column 5, line 66 to column 6, line 6), Eddy refers to other Viton polymers such as the copolymeric Viton A and the terpolymeric Viton B disclosed in Hartley. Throughout Eddy, these Viton polymers are consistently characterized as fluoroelastomers.

Kirk relates to synthetic thermoplastic elastomers, which, as discussed on page 16 of the reference, are typically multiphase systems containing thermodynamically incompatible mixtures of a hard polymer that becomes fluid on heating and a softer material that is rubberlike at room temperature. These two types of materials tend to separate into two phases, even when they are chemically combined into the same molecule by block or graft copolymerization. On page 18, Kirk states that "Not all thermoplastic elastomers are block copolymers. Those that are not are usually

combinations of a hard thermoplastic with a softer, more rubberlike polymer.

Usually, the components are mechanically mixed together, although it is sometimes possible to produce the rubber component *in situ* during polymerization." (emphasis added) Thus, Kirk teaches that thermoplastic elastomers are typically formed by the mechanical mixing, i.e., blending, of separate thermoplastic and elastomeric polymers.

Lewis includes definitions of "elastomer," "thermoplastic," and "thermoset" that can be summarized as follows:

An "elastomer" originally referred to "synthetic thermosetting high polymers having properties similar to those of vulcanized natural rubber..." when cross-linked; the " term was later extended to include uncross-linked polyolefins that are thermoplastic, ...whose extension and retraction properties are notably different from those of thermosetting elastomers..." (emphasis added) Thus, Lewis teaches that an "elastomer" thermoset by curing or cross-linking, as in the cured Viton elastomers employed in Hartley, Lentz, Schlueter, and Eddy et al., U.S. Patent No. 5,017,432, is a rubbery material.

The term "thermoplastic" refers to a "high polymer that softens when exposed to heat and returns to its original condition when returned to room temperature." Thermoplastic polymers include both natural substances such as crude rubber, i.e., rubber prior to its curing by vulcanization, and synthetic materials such as fluorocarbon polymers.

The term "thermoset " refers to a polymer that "solidifies or 'sets' irreversibly when heated," a property "usually associated with a cross-linking reaction of the molecular constituents induced by heat or radiation..." (emphasis added) The cross-linking reaction can be promoted by the addition of curing agents such as organic peroxides or (in the case of rubber) sulfur.

Based on the definitions provided by Lewis, the term "thermoset" applies both to cross-linked fluoroelastomers such as cured Viton A or Viton B and to cross-linked thermoplastic fluoropolymers such as cured THV fluorothermoplastics. Cured fluoroelastomers are thermosets; cross-linked thermoplastic fluoropolymers such as cured THV fluorothermoplastics are also thermosets, but they are not fluoroelastomers.

Blong is directed to a melt-processable coating composition that comprises a blend of a major amount of a melt-processable thermoplastic fluoropolymer

component and 0.01-20 wt.% of a poly(oxyalkylene) component. In the illustrative examples of Blong, 3M THV 500 Fluoroplastic is employed as the major thermoplastic fluoropolymer component, with Carbowax™ or Polyox™ polyethylene glycols being used as the poly(oxyalkylene) component. Thus, the coating compositions of Blong are blends of hard and soft polymeric components, as discussed in Kirk.

The thermoplastic elastomeric systems described by Kirk and exemplified in Blong are multiphase materials typically consisting of separate hard and soft components. By contrast, the applicants' fuser member includes a layer formed by curing of a coating composition containing a fluorocarbon thermoplastic random copolymer. It should be emphasized that this fluoropolymer is not only thermoplastic but also random, i.e., the reactant monomers are distributed substantially uniformly through a polymer formed in a single-step reaction. Such thermoplastic materials differ significantly from the various types of multiphase thermoplastic elastomers described in Kirk: the block or graft polymers that are formed in two or more reaction steps, and the blends, exemplified in Blong, that are produced by mixing two separately formed polymers.

As discussed in *Encyclopedia of Chemical Technology, Fourth Edition*, 1993, Volume 8, page 990, designated **Attachment A**, "Fluorocarbon elastomers are synthetic, noncrystalline polymers that exhibit elastomeric properties when cross-linked....In the 1960s, terpolymers of vinylidene fluoride, hexafluoropropylene, and tetrafluoroethylene, CF<sub>2</sub>=CF<sub>2</sub>, were developed and were commercialized by DuPont as Viton B." (emphasis added)

**Attachment B**, entitled "Comparison of Dupont Dow Viton® Fluoroelastomers," includes a description of Viton A as a fluoroelastomer dipolymer formed from vinylidene fluoride (VF2) and hexafluoropropylene (HFP) and Viton B as a fluoroelastomer terpolymer formed from vinylidene fluoride (VF2), hexafluoropropylene (HFP) and tetrafluoroethylene (TFE).

As described in Hull et al., "THV Fluoroplastic" in *Modern Fluoropolymers*, 1997, Chapter 13, pages 257-259, designated **Attachment C**, a melt-processable fluoroplastic consisting of tetrafluoroethylene (TFE), hexafluoropropylene (HFP), and vinylidene fluoride (VF2, as referred to as VDF) was developed by Hoechst AG in the early 1980s and is now marketed as Dyneon™ THV fluorothermoplastic. As mentioned on page 259, THV fluorothermoplastic is typically processed by melt

extrusion at temperatures in the range of 230-250°C. It should be noted that THV fluorothermoplastic is formed from the same three monomers as the fluoroelastomer Viton B.

**Attachment D**, entitled "Properties of Fluoroplastics," lists the material properties for two THV fluorothermoplastics, THV-400 and THV-500, which differ from one another primarily in their melting temperatures and specific gravities.

For purposes of completeness and convenience, copies of Attachments A, B, C, and D, which were previously discussed in the applicants' earlier filed Appeal Brief, are enclosed herewith.

Shifman et al., U.S. Patent No. 6,203,873 ("Shifman '873") discloses a blend of at least a first fluorinterpolymer characterized as a fluoroelastomer, and a second fluorinterpolymer characterized as a fluoroplastic, and also discloses that the first fluorinterpolymer having elastomeric characteristics and the second fluorinterpolymer having thermoplastic characteristics each comprises a copolymer, terpolymer, or mixture thereof formed by the copolymerization of two or more monomers selected from the group consisting of HFP, VF, and TFE.

Shifman '873 further discloses, at column 6, lines 24-29, that the blend used to form barrier layer 10, which is depicted in FIG. 1, contains a fluoroelastomer that can be a vinylidene fluoride-hexafluoropropylene-tetrafluoroethylene terpolymer such as a Fluorel® material, and a fluoroplastic that can be a tetrafluoroethylene-hexafluoropropylene-vinylidene fluoride terpolymer such as Dyneon THV.

Thus, Shifman '873, in disclosing Viton® and Fluorel® copolymers and terpolymers as useful commercial fluoroelastomers and Dyneon THV terpolymer as a useful commercial fluoroplastic, teaches that elastomeric and thermoplastic fluorinterpolymers, despite exhibiting markedly different physical characteristics, can both be formed from the same group of three monomers comprising the fluorocarbon thermoplastic random copolymer included in the coating composition of the present invention. Furthermore, it is clear that, based on the teachings of Kirk discussed above, that the blend of fluoroelastomer and fluoroplastic employed in the barrier layer of Shifman '873 constitutes a thermoplastic fluoroelastomer.

Shifman '873, at column 6, lines 31-43, discloses further embodiments, illustrated in FIGS. 2 and 3, that include an elastomeric inner tubular layer 12. The elastomer employed in layer 12 is selected from the "group consisting of nitrile rubber (NBR), thermoplastic fluoroelastomer, such as hexafluoropropylene vinylidene

copolymers or hexafluorenopropylene (*sic*)-vinylidene fluoride-tetrafluoroethylene terpolymers, polyvinyl chloride, and blends thereof. Preferably, the elastomeric, inner tubular layer is conductive NBR such as butadiene-acrylonitrile rubber." (emphasis added)

The foregoing quotation, which contains the only occurrence of the term "thermoplastic fluoroelastomer" in Shifman '873, also explicitly teaches that the elastomeric inner tubular layer 12, can be formed from a blend of polymers. As discussed above, the term "thermoplastic fluoroelastomer" characterizes the blend of a fluoroelastomer, for example, a vinylidene fluoride-hexafluoropropylene-tetrafluoroethylene terpolymer such as a Fluorel® material, and a fluoroplastic, for example, a tetrafluoroethylene-hexafluoropropylene-vinylidene fluoride terpolymer such as Dyneon THV, as is employed in the fabrication of barrier layer 10.

A closely related reference to Shifman '873 is Shifman, U.S. Patent Application Publication No. 2004/0187948 ("Shifman '948"), a copy of which is enclosed as **Attachment E**.

Shifman '948 discloses a flexible fuel hose having: a first barrier layer that contains a blend of a first fluorointerpolymer exhibiting elastomeric characteristics and having a fluorine content of about 68-74% and comprising a copolymer or terpolymer of two or more monomers selected from the group consisting of hexafluoropropylene, vinylidene fluoride and tetrafluoroethylene, and a second fluorointerpolymer exhibiting thermoplastic characteristics and having a fluorine content of about 73-78% and comprising a copolymer or terpolymer of two or more monomers selected from the group consisting of hexafluoropropylene, vinylidene fluoride and tetrafluoroethylene; a second fluoropolymer layer distinct from the first barrier layer and comprising a thermoplastic fluoropolymer of hexafluoropropylene, tetrafluoroethylene and vinylidene fluoride monomers; and a protective cover (page 1, [0006]; page 2, [0020], emphasis added).

It should be noted that the elastomeric first fluorointerpolymer and thermoplastic second fluorointerpolymer of the blend used to form the first barrier layer have overlapping fluorine contents and are each formed from the same group of three fluoromonomers. In addition, the second fluoropolymer layer, which is thermoplastic, is likewise formed from the same group of three fluoromonomers.

FIG. 3 of Shifman '948 is identical to FIG. 3 of Shifman '873. Shifman '948 discloses that, in the blend used to form barrier layer 10, the fluoroelastomer can be a

Viton or Fluorel® material, and the fluoroplastic can be Dyneon THV; this latter material can also be used to form an adjacent second barrier layer 12, which is distinct from the first barrier layer 10 (page 1, [0006]; page 3, [0032] and [0034], emphasis added). Thus, both Shifman '873 and Shifman '948 disclose a barrier layer 10 comprising a thermoplastic fluoroelastomeric blend. The hose depicted in FIG. 3 of Shifman '873 includes an elastomeric inner tubular layer 12, while that illustrated in FIG. 3 of Shifman '948 includes a fluorothermoplastic second barrier layer 12, which can be formed from Dyneon THV.

FIG. 4 of Shifman '948 discloses a further embodiment of the invention that includes, in addition to the first and second barrier layers 10 and 12, an elastomeric layer 14 adjacent to and surrounding the second barrier layer 12 (page 2, [0025]).

Thus, Shifman '948, like Shifman '873, discloses Viton® and Fluorel® copolymers and terpolymers as useful commercial fluoroelastomers and Dyneon THV terpolymer as a useful commercial fluoroplastic, confirming that elastomeric and thermoplastic fluorointerpolymers, despite exhibiting markedly different physical characteristics, can both be formed from the same group of three monomers. Shifman '948 also teaches that a barrier layer 10, formed from a thermoplastic fluoroelastomeric blend of a fluoroelastomer and a fluorothermoplastic, is distinct from a second barrier layer 12 formed from the same fluorothermoplastic alone.

In maintaining the §103(a) rejection of the claims in ¶¶4-5 of the Office Action mailed September 13, 2004, the Examiner twice asserted that "Although 'fluoroelastomer' Viton B of Hartley et al is substantially identical in structure and composition (emphasis added) to that of claimed fluorocarbon thermoplastic random copolymer, Hartley et al do not expressly state that 'fluoroelastomer' is thermoplastic." Hartley (as well as Lentz and Schlueter) do not state that a cured fluoroelastomer is thermoplastic because it is, in fact, not thermoplastic. In the course of this discussion, the applicants intend to provide irrefutable evidence that fluoroelastomers and fluorocarbon thermoplastic random copolymers, even if they are formed from the same monomers and are of similar chemical composition, are well recognized both in the patent prior art and in commercialized polymer technology as distinct classes of materials having substantially differing characteristics.

In ¶¶4-5 of the September 13, 2004 Office Action, the Examiner also twice asserted that "It is well known in the art that elastomers, including fluoroelastomers, include both thermosetting and thermoplastic polymers. See Lewis and Kirk in

entirety, especially page 25 of Kirk." Cured elastomers are thermosets, but the term elastomer alone is not understood in the art to include thermoplastic polymers. Kirk relates to synthetic thermoplastic elastomers, which are multiphase systems containing separate, incompatible soft (elastomeric) and hard (thermoplastic) components.

In ¶6 (A) of the September 13, 2004 Office Action, the Examiner stated that Lewis applies the term "thermoset" to crosslinked fluoroelastomers such as *cured* Viton B, and that the cured Viton B fluoroelastomer disclosed in Hartley is not thermoplastic. The applicants agree that cured Viton B is thermoset and not thermoplastic; they also agree with the Examiner's statements that their claim 1 recites that "a coating composition comprises a fluorocarbon thermoplastic random copolymer and a curing agent before curing" and that "Viton B in Hartley et al is cured *after* being coated onto the support as required by claim 1." However they respectfully disagree with the Examiner's assertion that the Viton B elastomer of Hartley is "embraced by the claimed fluorocarbon thermoplastic polymer because Viton B is uncured (non-crosslinked) elastomer having vinylidene fluoride, hexafluoropropylene and tetrafluoroethylene in proportions corresponding to those in the claimed fluorocarbon thermoplastic polymer." (emphasis added) It is well understood in the art that, as recited in the opening sentence of Attachment A, "Fluorocarbon elastomers are synthetic, noncrystalline polymers that exhibit elastomeric properties when crosslinked." (emphasis added) Commercially available elastomers such as Viton B are intended for use in their cured form in order to achieve their elastomeric properties. Various methods of curing fluoroelastomers such as Viton A and Viton B are extensively discussed in columns 3 and 4 of Hartley. Thermoplastic materials such as THV fluoropolymers, whether cured or uncured, do not exhibit elastomeric properties.

Both the terpolymeric Viton B and Viton A, a copolymer of hexafluoropropylene-vinylidene fluoride that is used in all the illustrative examples of Hartley, are cured, i.e., cross-linked in the coated layer, rendering them thermoset and elastomeric. By contrast, cured fluorocarbon thermoplastic polymers, although thermoset, are not fluoroelastomers.

In ¶6 (B) of the Office Action, the Examiner disagreed with the applicants' argument that the thermoplastic elastomer of Kirk is typically a multiphase block copolymer or a mechanically blended mixture, stating that it "is sometimes possible to

produce the rubber component *in situ* during polymerization (i.e. forming a random copolymer)." In response, the applicants respectfully point out that thermoplastic elastomers, no matter what multistep procedure is used to form them, are multiphase polymeric systems that include distinct hard and soft phases and are thus dissimilar from the cured fluorocarbon thermoplastic random copolymer of the applicants' invention.

The differing positions of the Examiner and the applicants appear to stem in large measure from a disagreement as to the meaning of the terms "elastomer," "thermoplastic," "thermoset," and "thermoplastic elastomer," and the differentiation of these classes of materials. In undertaking to resolve this disagreement, the applicants offer a two-page excerpt from the O-Ring Catalog of the Simrit/Freudenberg-NOK Company ("Simrit"), a copy of which is enclosed as **Attachment F**.

Simrit characterizes a "thermoplastic" as a polymer that can be melted with the application of heat. The more general term "plastic" includes both "thermoplastic" and "thermoset" materials, the latter undergoing a change in chemical structure and properties but not melting with the application of heat. "As a class, plastics have low elongation and high elongation set." Simrit further describes an "elastomer" as being a flexible long- chain polymer that is capable of cross-linking, resulting in the "elastic, or rubbery, properties of these materials." (emphasis added)

A "thermoplastic elastomer," or "TPE," as described in Simrit, combines the properties of a an elastomer with the ease of processability of a thermoplastic, "the result of a physical combination of soft, elastic polymer segments and hard, crystalline segments which are capable of crosslinking. Thermoplastic elastomers are generally classified by their structure rather than their chemical makeup." (emphasis added) This description of thermoplastic elastomers is consistent with that presented in greater detail in Kirk, discussed above.

As already noted, Viton® and Fluorel® materials are examples of commercially available fluoroelastomers, and THV materials are well-known commercially available fluoroplastics. Thermoplastic fluoroelastomers are also items of commerce, one example being DAI-EL Thermoplastic from Daikin Industries, Ltd. This material, which consists of a soft fluoroelastomer segment connected to a hard fluororesin segment, is schematically illustrated in enclosed **Attachment G**.

Hartley discloses curing a coated fluoroelastomer-containing layer at temperatures of at least 230°C; similarly, Lentz includes an example with a curing

temperature of 232°C. Neither Hartley nor Lentz nor any of the other cited references teach the use of a metal oxide or combination of oxides to enable low temperature curing of a fluoropolymer layer. By contrast, the applicants' method of making a fuser advantageously provides for the low-temperature curing, at a temperature of 25°C to 120°C, preferably 25°C to 50°C, more preferably 25°C, of a layer formed from a composition containing a fluorocarbon thermoplastic random copolymer such as THV, along with antimony-doped tin oxide particles.

Effenberger et al., U.S. Patent No. 5,194,335 ("Effenberger"), cited in the July 17, 2002 Office Action, teaches a fluoroplastic resin dispersion modified by the addition of a fluoroelastomer latex (Abstract, emphasis added), thereby providing further evidence that fluoroelastomers and fluoroplastics are understood by those skilled in the art to be distinctly different classes of materials, which, however, can be combined to form multiphase thermoplastic fluoroelastomers. Columns 3 and 4 of this patent contain separate lists of suitable fluoroplastics and fluoroelastomers to be combined in the disclosed coating compositions; Viton is included in the list of preferred fluoroelastomers.

In support of the request for withdrawal of the rejections of the claims and the allowance of this application, the applicants respectfully call to the Examiner's attention the following references that differentiate among the various types of fluoropolymeric materials:

Chmielewski, U.S. Patent No. 6,624,251 (Chmielewski"), a copy of which is enclosed as **Attachment H**, discloses a two-phase fluorinated polymeric composition that is useful as a thermoplastic vulcanizate and includes a continuous phase of at least one thermoplastic fluorocarbon resin and a disperse phase that is a blend of two or more fluoroelastomers, at least one of which is a vinylidene fluoride-tetrafluoroethylene-propylene terpolymer (Abstract). Suitable fluorothermoplastics include Dyneon THV materials (column 3, lines 43-47); suitable fluorelastomers include DuPont-Dow Viton terpolymeric materials (column 4, lines 31-33).

Fukushi et al., U.S. Patent No. 6,773,755 ("Fukushi"), a copy of which is enclosed as **Attachment J**, discloses a process for forming a multilayered article having a fluoroplastic layer and an elastomer layer that can comprise a fluoroelastomer. The background section of the reference contains a description of fluoropolymers, including cured or crosslinked fluoroelastomers and semi-crystalline or glassy fluoroplastics (column 1, lines 17-42). Useful commercial fluoroplastics for

the fluoroplastic layer of the article include THV fluoropolymers THV 200, THV 400, THV 500G and THV 610X from Dyneon (column 3, lines 48-51), and suitable fluoroelastomers for the elastomer layer include VDF-HFP copolymers and VDF-HFP-TFE terpolymers (column 3, lines 62-64). As noted in **Attachments C** and **B**, both THV fluoroplastics and Viton® B fluoroelastomer are terpolymers prepared from the VDF, HFP and TFE monomers.

Rees, U.S. Patent No. 5,006,594, ("Rees"), a copy of which is enclosed as **Attachment K**, discloses a fluorinated thermoplastic elastomeric blend that contains a two-phase composition including a continuous phase of a melt-processible crystalline thermoplastic fluorocarbon resin and a dispersed phase of an amorphous crosslinked fluoroelastomer, together with a non-melt-processible polytetrafluoroethylene powder (Abstract). The thermoplastic fluorocarbon resin in the continuous phase contains a minimum of 38 wt.% fluorine, and the fluoroelastomer in the dispersed phase contains a minimum of 50 wt.% fluorine (column 2, lines 29-37). The reference discloses that fluoroelastomers useful in the disclosed compositions are well known and usually commercially available, including copolymers of vinylidene fluoride and one or more fluoromonomers containing 2 to 8 carbon atoms, for example, copolymers of vinylidene fluoride and hexafluoropropylene (column 3, lines 22-28). Useful thermoplastic polymers also include copolymers of vinylidene fluoride and hexafluoropropylene (column 3, lines 9-13). Thus both the fluoroelastomeric and the fluoroplastic components of the blend can be formed from the same fluoromonomers, and the fluorine content of the fluoroelastomer overlaps that of the fluoroplastic.

Kaduk et al., U.S. Patent No. 5,741,855 ("Kaduk"), a copy of which is enclosed as **Attachment L**, discloses a composition that is a blend of a first crystalline fluoroplastic that is an ethylene-tetrafluoroethylene copolymer and a second crystalline fluoroplastic that is a tetrafluoroethylene-vinylidene fluoride-hexapropylene terpolymer, together with a compatibilizer that is a thermoplastic fluoroelastomer comprising an elastomeric and a nonelastomeric segment (column 2, lines 2-26). A suitable second crystalline fluoroplastic is commercially available under the tradename THV from 3M Company (column 2, line 66, to column 3, line 2), and a preferred thermoplastic fluoroelastomer is available from Daikin under the tradename Daiel T-530 (column 3, lines 29-30). Thus, this reference teaches a fluoropolymeric composition that can include both a commercial fluoroplastic, THV, and a commercial thermoplastic fluoroelastomer, Daiel T-530.

Johnson et al., U.S. Patent No. 5,275,887 ("Johnson"), a copy of which is enclosed as **Attachment M**, discloses a composition comprising a blend of (a) a thermoplastic copolymer of vinylidene fluoride and hexafluoropropylene and (b) a fluoroelastomer copolymer whose repeating units contain about 30-70 mole percent vinylidene fluoride and about 70-30 mole percent hexafluoropropylene (cf. claim 1, emphasis added). Thus, this reference teaches a composition that is a blend of a thermoplastic fluoropolymer and a fluoroelastomer, both formed from the same monomers.

Faulkner, U.S. Patent No. 6,538,069 ("Faulkner"), a copy of which is enclosed as **Attachment N**, discloses a moldable, extrudable thermally crosslinkable composition of matter containing 50-99 wt.% fluoropolymers, in which 50-95 % of the polymer content of the blend is an FKM fluoroelastomer and 5-50 % of the polymer content of the blend is one or more thermoplastic PVDF polymers or copolymers containing at least 70 wt.% vinylidene fluoride monomer units (Abstract). Viton A from DuPont is mentioned as commercially available FKM fluoroelastomer (column 2, lines 30-35), and reference is made to rubber/plastic blends of FKM with THV polymers from Dyneon as being known in the prior art (column 1, lines 28-29).

Novak et al., U.S. Patent No. 5,371,143 ("Novak"), a copy of which is enclosed as **Attachment O**, discloses a fluorine- containing thermoplastic elastomeric composition comprising a continuous phase having a melt temperature or glass transition temperature greater than about 150°C, a block thermoplastic elastomer having a melt temperature greater than about 150°C, and a disperse phase comprising particles or domains of cured fluoroelastomer composition (Abstract). The reference teaches that thermoplastic elastomers are materials that possess both elastomeric and thermoplastic properties and are of two main types: block and graft copolymers containing elastomeric and plastic polymer chain segments, and blends of certain elastomers and thermoplastics (column 1, lines 58-66). Commercially available fluoroelastomers useful in the disperse phase include FLUOREL and VITON materials, among others (column 7, lines 15-19).

The foregoing discussion of the cited references and attachments can be summarized as follows:

- 1) Fluoroelastomers and fluorothermoplastics are recognized in the patent prior art and in commercialized polymer technology as distinct classes of materials having substantially differing characteristics.

2) Known commercial fluoroelastomeric materials include Viton® A and Viton® B, whose cured forms are disclosed as useful fluoroelastomers for the compositions of Hartley, as well as the compositions of Lentz, Schlueter, and Eddy.

3) Known commercial fluorothermoplastics include THV materials, an example of which, THV 200A, is employed in the illustrative examples of the composition of the present invention.

4) Although a fluoroelastomer and a fluorothermoplastic differ in their physical characteristics, they can be formed from the same group of monomers, for example, VDF, HFP and TFE, which are the monomeric components, in differing but overlapping weight ratios, of both a THV fluoroplastic and Viton® B fluoroelastomer.

5) Curing or crosslinking of a fluoroelastomer and a fluoroplastic both result in the formation of a thermoset polymer; the cured fluorelastomer, however, is elastic, while the cured fluoroplastic is rigid.

6) A thermoplastic fluoroelastomer, which can be produced by blending a separately formed fluoroelastomer with a separately formed fluorothermoplastic or by a block or graft polymerization is a multiphase polymeric system that includes distinct hard and soft phases, as described in Kirk and in **Attachments H, K, M, and O.**

7) A commercially available example of a multiphase thermoplastic fluoroelastomer that is a thermodynamically incompatible mixture of a hard polymer and a softer rubberlike polymer is DAI-EL Thermoplastic.

The applicants again respectfully call attention to the following eight recent patents, each of which issued from an application filed June 30, 2000 and, with one exception, was each allowed by a different primary examiner:

U.S. Patent Nos. 6,355,352; 6,361,829; 6,372,833; 6,416,819; 6,419,615; 6,429,249; 6,444,741; and 6,696,158.

Each of these eight patents discloses and claims a coating composition that comprises a fluorocarbon thermoplastic random copolymer containing the same ranges of the same monomers as that recited in claim 1 of the instant application, Serial No. 09/608,818, also filed June 30, 2000, i.e., subunits of  $-(CH_2CF_2)_x-$ ,  $-(CF_2CF(CF_3)_y-$ , and  $-(CF_2CF_2)_z-$ , wherein x is from 1 to 50 or 60 to 80 mole

percent, y is from 10 to 90 mole percent, z is from 10 to 90 mole percent, and x + y + z equals 100 mole percent.

Hartley, the lead reference in the rejection of the instant application, is a cited reference in every one of these patents except U.S. Patent No. 6,419,615. The issuance of the aforementioned eight related patents disclosing compositions containing fluorothermoplastics in light of cited prior art disclosing compositions containing fluoroelastomers provides further convincing evidence that these two types of fluoropolymers are recognizably different and patently distinct materials.

Because the combined teachings of the cited prior art references fail to render obvious the applicants' invention, withdrawal of the §103(a) final rejections of the claims is respectfully requested. Claims 1-5 and 7-22 are now in this case, whose prompt allowance is earnestly solicited.

Respectfully submitted,

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Enclosures: Attachments A, B, C, D, E, F, G, H, J, K, L M, N, O

Attachment A  
*Encyclopedia of Chemical Technology, Fourth Edition, Volume 8*

990      ELASTOMERS, SYNTHETIC (FLUOROCARBON)

Vol. 8

### FLUOROCARBON ELASTOMERS

Fluorocarbon elastomers are synthetic, noncrystalline polymers that exhibit elastomeric properties when cross-linked. They are designed for demanding service applications in hostile environments characterized by broad temperature ranges and/or contact with chemicals, oils, or fuels.

Military interest in the development of fuel and thermal resistant elastomers for low temperature service created a need for fluorinated elastomers. In the early 1950s, the M. W. Kellogg Co. in a joint project with the U.S. Army Quartermaster Corps, and 3M in a joint project with the U.S. Air Force, developed two commercial fluorocarbon elastomers. The copolymers of vinylidene fluoride,  $\text{CF}_2=\text{CH}_2$ , and chlorotrifluoroethylene,  $\text{CF}_2=\text{CFCl}$ , became available from Kellogg in 1955 under the trademark of Kel-F (1-3) (see **FLUORINE COMPOUNDS, ORGANIC—POLYCHLOROTRIFLUOROETHYLENE; POLY(VINYLIDENE) FLUORIDE**). In 1956, 3M introduced a polymer based on poly(1,1-dihydroperfluorobutyl acrylate) trademarked 3M Brand Fluororubber 1F4 (4). The poor balance of acid, steam, and heat resistance of the latter elastomer limited its commercial use.

In the late 1950s, the copolymers of vinylidene fluoride and hexafluoropropylene,  $\text{CF}_2=\text{CFCF}_3$ , were developed on a commercial scale by 3M (Fluorel) and by Du Pont (Viton) (5-8). In the 1960s, terpolymers of vinylidene fluoride, hexafluoropropylene, and tetrafluoroethylene,  $\text{CF}_2=\text{CF}_2$ , were developed (9) and were commercialized by Du Pont as Viton B. At about the same time, Montedison developed copolymers of vinylidene fluoride and 1-hydropentafluoropropylene as well as terpolymers of these monomers with tetrafluoroethylene, marketed as Tecnoflon polymers (10,11).

In the 1960s and 1970s, additional elastomers were developed by Du Pont under the Viton and Kalrez trademarks for improved low temperature and chemical resistance properties using perfluoro(methyl vinyl ether),  $\text{CF}_2=\text{CFOCF}_3$ , as a comonomer with vinylidene fluoride and/or tetrafluoroethylene (12,13) (see **FLUORINE COMPOUNDS, ORGANIC—TETRAFLUOROETHYLENE POLYMERS AND COPOLYMERS**).

Bromine- and iodine-containing fluoroolefins have been copolymerized with the above monomers in order to allow peroxide cure (14-21). The peroxide cure system does not require dehydrofluorination of the polymer backbone, resulting in an elastomer that shows improved properties after heat and fluid aging.

Copolymers of propylene and tetrafluoroethylene, which are sold under the Aflas trademark by 3M, have been added to the fluorocarbon elastomer family (21-26). Also 3M has introduced an incorporated cure copolymer of vinylidene fluoride, tetrafluoroethylene and propylene under the trademark Fluorel II (27). These two polymers (Aflas and Fluorel II) do not contain hexafluoropropylene. The substitution of hexafluoropropylene with propylene is the main reason why these polymers show excellent resistance toward high pH environments (28). Table 1 lists the principal commercial fluorocarbon elastomers in 1993.

### Properties

Table 2 summarizes general characteristics of vulcanizates prepared from commercially available fluorocarbon elastomer gumstocks.




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## Comparison of Dupont Dow Viton® Fluoroelastomers

There are three major general use grades of Viton® fluoroelastomer: A, B and F. They differ primarily in their resistance to fluids, and in particular aggressive lubricating oils and oxygenated fuels, such as methanol and ethanol automotive fuel blends. There is also a class of high performance Viton® grades: GB, GBL, GP, GLT, and GFLT.

### General Use Grades:

#### **Viton® A: (vinylidene fluoride and hexafluoropropylene)**

Viton® A is a family of fluoroelastomer dipolymers, that is they are polymerized from two monomers, vinylidene fluoride (VF2) and hexafluoropropylene (HFP). Viton® A fluoroelastomers are general purpose types that are suited for general molded goods such as o-rings and v-rings, gaskets, and other simple and complex shapes. There is a full range of Viton® A grades that accomodate various manufacturing processes including transfer and injection molding, extrusion, compression molding, and calendering.

#### **Viton® B: (vinylidene, hexafluoropropylene and tetrafluoroethylene)**

Viton® B is a grade of fluoroelastomer terpolymers, that is they are polymerized from three monomers, vinylidene (VF2), hexafluoropropylene (HFP), and tetrafluoroethylene (TFE). Viton® B

fluoroelastomers offer better fluid resistance than A type fluoroelastomer. There is a full range of Viton® B grades that accommodate a variety of manufacturing processes including injection and compression molding, extrusion, and calendering.

#### **Viton® F:**

Viton® F is a grade of fluoroelastomer terpolymers, that is they are polymerized from three monomers, vinyl fluoride (VF2), hexafluoropropylene (HFP), and tetrafluoroethylene (TFE). Viton® F fluoroelastomers offer the best fluid resistance of all Viton® types. F types are particularly useful in applications requiring resistance to fuel permeation. There is a range of Viton® F grades to accommodate various manufacturing requirements.

#### **High Performance Grade:**

#### **Viton® GB, GBL:**

Viton® GB and GBL are grades of fluoroelastomer terpolymers, that is they are polymerized from three monomers, vinyl fluoride (VF2), hexafluoropropylene (HFP), and tetrafluoroethylene (TFE). Viton® GB and GBL use peroxide cure chemistry that result in superior resistance to steam, acid, and aggressive engine oils. There is full range of GB and GBL types that can accommodate most rubber processing requirements including compression, injection and transfer molding, extrusion, and calendering.

#### **Viton® GLT:**

Viton® GLT is a fluoroelastomer designed to retain the high heat and the chemical resistance of general use grades of Viton® fluoroelastomer, while improving the low temperature flexibility of the material. Glass transition temperatures (Tg) of materials are indicative of low temperature performance in typical elastomer applications. Viton® GLT shows an 8 to 12°C lower Tg than general use Viton® grades. There is a range of GLT products to accommodate various processing conditions.

# 13

## THV Fluoroplastic

D. E. HULL, B. V. JOHNSON, I. P. RODRICKS and  
J. B. STALEY

Fluorothermoplastic Business Unit, Dyneon, St Paul, MN, USA

### 1 BACKGROUND

In the early 1980s Hoechst AG developed a commercial production process for a unique melt processable fluoroplastic consisting of tetrafluoroethylene, hexafluoropropylene, and vinylidene fluoride (Figure 13.1). The impetus for this product was a contract to develop an outdoor fabric coating that could provide the protection of typical fluoroplastics like PTFE and ETFE, but additionally could be used with PVC-coated polyester fabric without significantly compromising overall flexibility. PTFE and ETFE could not be used since their high melt fusion temperatures destroyed PVC-coated polyester fabric. Hoechst developed the product and conducted a restricted test market (primarily fabric coating within Germany) under the brandname Hostafon™ TFB X. In January 1993 Hoechst granted worldwide marketing rights to 3M and the product reached full commercial status under the name 3M™ THV Fluoroplastic. With the start-up of the Dyneon 3M/Hoechst joint venture in August 1996, the product is now known as Dyneon™ THV fluorothermoplastic.

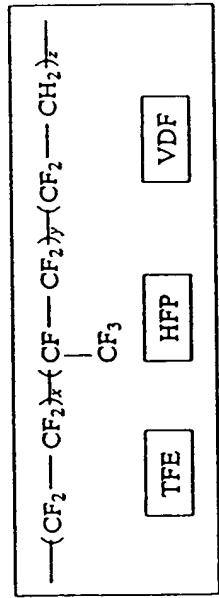


Figure 13.1. Chemical structure of THV terpolymer. (Reproduced with permission)

## 2 MANUFACTURING

THV Fluoroplastic is produced in Gendorf, Germany. THV Fluoroplastic is polymerized under aqueous emulsion conditions and the resulting dispersions can be sold and used directly or following concentration with an emulsifier. Dry forms result from coagulation, washing, filtering and drying of the dispersion followed by extrusion (pellets) or grinding (powder) operations. Additives are generally not added to THV Fluoroplastic since the product is inherently very stable and easy to process. An electrostatic dissipative compound has been developed and was commercialized in 1995.

Products are available in 25 kilogram bags or larger containers (pellets), 50 kilogram drums (powders) or 35 kilogram (wet weight dispersions) barrels. No chlorofluorocarbons are employed at any step during the production process and the relatively recent construction of the production facilities allowed implementation of state of the art technology.

## 3 PROPERTIES

In general, THV Fluoroplastic is set apart from other melt-processable fluoroplastics by a combination of properties that include relatively low processing temperatures; bondability (to itself and other substrates); high flexibility; excellent clarity and low refractive index; and efficient E-beam crosslinking. One grade, THV-200, is soluble in common organic solvents. THV Fluoroplastic also retains properties common to other commercial fluoroplastics such as chemical resistance, weatherability, and low flammability.

There are four commercial THV Fluoroplastic grades (three dry and one aqueous dispersion) differing in the monomer ratios that subsequently influence melting points, chemical resistance and flexibility. Table 13.1 shows the basic properties of the three dry grades. THV-200 has the lowest melting point, the least chemical resistance of the THV Fluoroplastic grades (soluble in common solvents such as ethyl acetate, ketones, etc.), is the easiest to E-beam crosslink and is the most flexible. THV-500 has the best chemical and permeation resistance of the THV Fluoroplastic grades. THV-400 has a slightly lower melting point than THV-500 to meet specific processing requirements.

THV Fluoroplastic is available in pellet (THV-200G, 400G and 500G); powder (THV-200P) and aqueous dispersions (THV-330R 30% solids and 350C 50% solids).

THV Fluoroplastic is processed within the fluoroplastics fabricator base, but because of its relatively low processing temperature it is also processed by olefinic processors. Virtually all of the melt-processable plastic processing methods are used with THV Fluoroplastic including extrusion, co-extrusion and blow molding) with plastics as well as with various elastomers.

**Table 13.1. Typical THV properties (nominal values, not for specification purposes). (Reproduced with permission)**

Property	ASTM method	THV grade		
		THV 200	THV 400	THV 500
Specific gravity	D792	1.95	1.97	1.98
Melting range (°C)	D3418	115-125	150-160	165-180
Thermal decomposition in air (°C)	TGA	420	430	440
Limiting oxygen index (LOI)	D2863	65	NA	75
Tensile strength at break (psi)	D638*	4200	4100	4100
Tensile strength at break (MPa)	D638*	29.0	28.3	28.3
Elongation at break	D638*	600%	500%	500%
Flexural modulus (psi)	D790	12,000	NA	30,000
Flexural modulus (MPa)	D790	82.7	NA	206.7
Hardness, Shore D	D2240	44	53	54
Dielectric constant at 23 °C				
100 kHz	D149	6.6	5.9	5.6
10 mHz	D149	4.6	4.1	3.9
Melt flow index (gm/10 min @ 260 °C 5 kg)	D1238	20	10	10
E-beam cured high temperature Resistance (°C)	NA	>150	NA	NA

\*Property measured on extruded film

tandem extrusion; blow and coblow molding (including blown film); injection molding, skived film; vacuum forming; and solvent casting (only for THV-200). These processes produce film, tubing, containers, profiles and molded shapes. Many of these products involve multilayer constructions where THV Fluoroplastic provides chemical, barrier or other properties in a relatively thin layer bonded to thicker layers of structural plastics or elastomers of various types.

### 3.1 THV FLUOROPLASTIC DISTINGUISHING FEATURES

As mentioned earlier, THV Fluoroplastic has a unique combination of properties that set it apart from other melt-processable fluoroplastics. These features are beneficial not only to end-users but to manufacturers faced with part production and design challenges.

#### 3.1.1 THV Fluoroplastic Processing Temperatures

During most extrusion processing, THV Fluoroplastic melt temperature at the die is in the 230-250 °C range. This relatively low processing temperature presents many new options for co-processing (e.g., coextrusion, cross-head extrusion, co-blow molding) with plastics as well as with various elastomers.

Attachment D

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## Fluoroplastics

State: THV-400

## Material Properties

### Mechanical Properties

	Conditions	
	State 1	State 2
Tensile Strength (MPa)	23	at break
Elongation at break (%)	500	
Hardness	53	Shore

### Physical & Electrical Properties

	Cond
	State
Specific Gravity	1.97

### Processing Properties

	Conditions	
	Type	
Melting Temperature (°C)	150	T <sub>m</sub> , crystalline
Processing Temperature (°C)	243	extrusion

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### Fluoroplastics

State: THV-500

### Material Properties

#### Mechanical Properties

		Conditions	
		State 1	State 2
Flexural Modulus (MPa)	207	23 °C	
Tensile Strength (MPa)	23	at break	
Elongation at break (%)	500		
Hardness	54	Shore	

#### Physical & Electrical Properties

Cond  
State

Specific Gravity

1.78

#### Processing Properties

Conditions  
Type

Melting Temperature (°C)

180

T<sub>m</sub>, crystalline

Processing Temperature (°C)

249

extrusion

### Suppliers

Dyneon

Oakdale, MN 800-863-9374

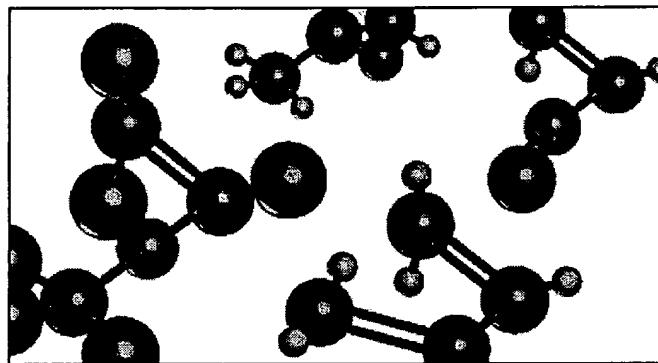
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Copper, Titanium

## POLYMER HISTORY

As far back as 1839, Charles Goodyear first improved the elastic properties of natural rubber by heating with sulfur (vulcanization). It was not until the 1930s that the macromolecule model of rubber was understood. After World War II and through the 1950s rapid developments in synthetic polymers were made. Most commercial high-performance elastomers trace their origins to the 1960s and 1970s.

Polymers are long chains of repeating chemical units, or monomers. The chemical skeletal structures may be linear, cyclic or branched. When one monomer is polymerized, the resultant polymer is called a homopolymer. Examples include polyethylene, polystyrene and polytetrafluoroethylene (PTFE). Copolymers (or dipolymers) are derived from the polymerization of more than one type of monomer. The distribution of monomers in these copolymers can be statistical, random or alternating. Examples include ethylene - propylene and fluorocarbon elastomers (vinylidene fluoride and hexafluoropropylene). Terpolymers are three - monomer - unit polymers, such as ethylene - propylene - diene (EPDM) and specialty fluorocarbon grades.



## TYPES OF POLYMERS

There are three general classes of polymers:

1. **Thermoplastics** (can be melted with the application of heat)

- **Crystalline**— crystallize when cooled
- **Amorphous**— no crystallization when cooled
- **Semicrystalline**— polymers which contain both crystalline and amorphous segments

2. **Thermosets** (degrade rather than melt with the application of heat)

### 3. Elastomers (cross-linked)

**Plastics** are rigid long-chain polymers which are not usually connected or cross-linked. Plastics can either be thermoplastic—meaning they can be heated and cooled without changing properties—or thermoset, where an increase in temperature changes the chemical structure and properties. As a class, plastics have low elongation and high elongation set.

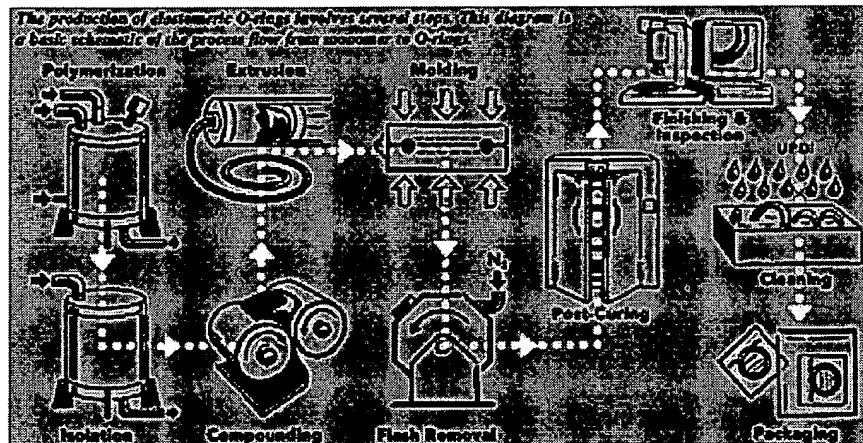


**Elastomers** are flexible long-chain polymers which are capable of cross-linking. Cross-linking chemically bonds polymer chains which can prevent reversion to a non-cross-linked polymer at elevated temperatures. The cross-link is the key to the elastic, or rubbery, properties of these materials. The elasticity provides resiliency in sealing applications.

**Thermoplastic** elastomers (TPEs) often combine the properties of elastomers with the ease of processability of thermoplastics. They are the result of a physical combination of soft, elastic polymer segments and hard, crystalline segments which are capable of cross-linking. Thermoplastic elastomers are generally classified by their structure rather than their chemical makeup.

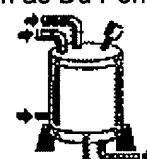


## RUBBER PROCESSING



### Polymerization

The beginning step for elastomers is the polymerization of the backbone and cure-site monomers. This is typically done by large chemical companies such as Du Pont, Dow, GE, Ausimont, Daikin and Dyneon. Common techniques are emulsion, microemulsion, and suspension polymerization.



Polymerization combines two or more process gases (monomers) into an aqueous environment and under specific temperature and pressure conditions connects the individual monomers into the desired polymer.

Initiating agents, buffers and other chemicals may be added to the polymer reactor to achieve the desired chemical properties and polymerization dynamics.

### Isolation

The backbone polymers are isolated (brought out of the emulsion), cleaned and dried. Chemical agents may be added at this step to isolate the polymer "latex" into

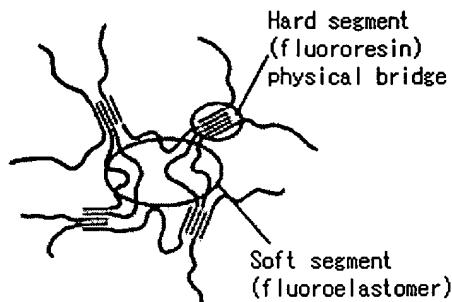




## ■ Thermoplastic fluoroelastomer DAI-EL Thermoplastic

[BACK](#)

DAI-EL Thermoplastic is a polymer consisting of fluororesin and fluoroelastomer. A unique feature of DAI-EL thermoplastic is that it can be formed like a thermoplastic while having properties similar to those of a fluoroelastomer.



DAI-EL Thermoplastic Concept Drawing

### ■ Features

- Good transparency
- Excellent chemical resistance, Low extractable substance
- Recyclable



Application example

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